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Soliton Generation in Semi-Infinite Molecular Chains

By

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A mechanism for generating solitons in semi-infinite molecular chains by exciting the impurity molecules is considered. The generation criteria of solitons are found and their dependence on molecular chain parameters, the input place, and localization degree of initiating excitation. The possibility to govern the generation efficiency and solitons velocity by changing the parameters of exciting signal is pointed out.

Рассмотрен механизм генерации солитонов в полубесконечных молекулярных цепочках путем возбуждения примесных молекул. Найдены критерии генерации солитонов и их зависимость от параметров молекулярной цепочки, места подачи и степени локализации инициирующего возбуждения. Указано на принципиальную возможность управления эффективностью генерации и скоростью солитонов посредством изменения параметров возбуждающего сигнала.

1. Introduction

The perspectives to use the soliton mechanism of energy and charge transport in transferring information in quasi-one-dimensional molecular structures [1 to 4] have intensively been discussed in recent years. Special attention is paid to the soliton generation problem as its solution makes it possible to control the efficiency of soliton generation, its velocity, characteristic size, and amplitude.

The soliton generation problem has two aspects: 1. to create the necessary initial distribution of excitations of the required nature, 2. to form a soliton on the basis of this distribution. These aspects are intimately related; however, under certain conditions they can be considered independently and the soliton generation problem can be solved stage by stage. Such an approach is possible e.g. in the case when the times necessary for creating the initial distribution are sufficiently small so that the mechanisms responsible for the redistribution of excitations between molecules are not able to operate.

In terms of the above-cited treatment it has been managed to study in some detail the second aspect of the problem and to show that a soliton is most effectively formed using the initial distribution of some optimal size which is given by the chain parameters [1, 5, 6]. Being quite appreciable by itself the above-cited result proves to be an argument in favour of the impossibility of direct soliton generation due to light [1, 3] since the relative smallness of the optimal size restricts the admitted wavelengths of the exciting radiation.

In this connection the possible mechanism for soliton generation turns out to be the following: 1. electron, light quantum, or another excitation capture by the impurity molecule (chromophore, for example) coupled with the principal chain in a nonresonant way; 2. fast excitation transfer to neighbouring molecules of the principal chain (crea-

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tion of a localized initial distribution); 3. soliton formation using the initial distribution obtained in such a way. If along the principal chain one disposes different impurities (chromophores), this makes it possible to change the location of the signal input to the principal chain and/or its localization degree (e.g., by changing the frequency and/or the exciting light frequency detuning, respectively) and, thus, to govern the soliton generation parameters.

2. Equations for the Evolution of the Excitation

In solving the soliton generation problem it is tempting to make use of the inverse scattering problem for some exactly solvable model. So the problem of soliton generation in molecular chains can be reduced to considering the nonlinear Schrödinger equation. However, this is possible only at small inertia of the elastic subsystem [7],

$$4MJ^2/wh^2 \ll 1 , \tag{1}$$

where M is the molecule mass, J the resonance excitation transfer constant, w the elasticity coefficient. We shall elucidate how such a program can be realized in semi-infinite molecular chains exploiting the soliton generation through the impurity connected with the s-th molecule of the principal chain.

If one excites the impurity molecule in a certain way the further redistribution of this excitation over the chain in the mean field approximation is described by a system of equations

$$ih\frac{\mathrm{d}}{\mathrm{d}t}\varrho_{nm} = i\Gamma\delta_{ns}\delta_{sm}R + J[(1-\delta_{n1})\varrho_{n-1m} + \varrho_{n+1m} - (1-\delta_{1m})\varrho_{nm-1} - \varrho_{nm+1}] +$$

$$+ \frac{\chi^2}{w}[\varrho_{n+1\,n+1} + \varrho_{nn} + (1-\delta_{n1})(\varrho_{nn} + \varrho_{n-1\,n-1}) - \varrho_{m+1\,m+1} -$$

$$- \varrho_{mm} - (1-\delta_{1m})(\varrho_{mm} + \varrho_{m-1\,m-1})]\varrho_{nm} , \tag{2a}$$

$$h\frac{\mathrm{d}}{\mathrm{d}t}R = -\Gamma R , \tag{2b}$$

$$\varrho_{nm}(t=0) = 0 \ (n, m = 1, 2, 3, \dots, \infty) , \ R(t=0) = 1 \tag{2c}$$

and the longitudinal displacements of molecules of the principal chain β_n, in virtue of (1), are connected with the values ϱ_{nn} by the relation

$$\beta_n = \frac{\chi}{w}\sum_{m=n}^{\infty}(\varrho_{mm} + \varrho_{m+1\,m+1}) \qquad (n = 1, 2, 3, \dots, \infty) . \tag{3}$$

Here ϱ_{nn} is the probability of the presence of excitation on the n-th molecule, R the probability of the presence of excitation on the impurity, Γ the constant of nonresonant excitation transfer from the impurity molecule to a neighbouring one of the basic substance, χ the interaction constant of an excitation with displacements in the principal chain. Equations (2a) to (2b) are written under the assumption that the resonance excitation transfer between the impurity and the host molecules and also the probability of the thermal excitation to the impurity are negligibly small. This is justified when the difference in the excited state energies of impurity molecules and the basic substance is sufficiently great.

To realize the procedure cited in the Introduction of distinguishing two problems out of the initial one ((2a) to (2b)) it is necessary to require the fulfilment of the condition

$$\Gamma \gg J. \tag{4}$$

Then the problem of creating the initial distribution is solved easily, i.e. at a certain time moment t_0, such that

$$h/\Gamma \ll t_0 \ll h/J \tag{5}$$

the excitation appears to be almost localized on the molecule neighbouring the impurity,

$$\varrho_{nm}(t = t_0) \approx \delta_{ns}\delta_{sm}, \tag{6}$$

and the reverse transition of excitation to the impurity is impossible

$$R(t \gtrsim t_0) = 0. \tag{7}$$

Besides in this case at time $t \gtrsim t_0$ in (2a) the decoupling

$$\varrho_{nm} = \frac{2}{g} r_n r_m^* \equiv \frac{2}{g} r_n q_m \tag{8}$$

takes place, where

$$g = 4\chi^2/wJ. \tag{9}$$

Thus the problem of soliton formation on the basis of the initial distribution (6) can be formulated in terms of the probability amplitudes

$$i \frac{d}{d\tau} r_n - r_{n+1} - (1 - \delta_{n1}) r_{n-1} + 2r_n -$$

$$- \frac{1}{2} [r_{n-1}q_{n-1} + r_n q_n + (1 - \delta_{n1}) (r_n q_n + r_{n-1}q_{n-1})]r_n = 0, \tag{10a}$$

$$r_n(\tau = 0) = \sqrt{\frac{g}{2}} \delta_{ns} \qquad (n = 1, 2, 3, \dots, \infty), \tag{10b}$$

where

$$\tau = J(t - t_0)/h. \tag{11}$$

3. Soliton Formation

It is convenient to consider instead of the problem (10a) to (10b) on the semi-infinite chain the following one:

$$i \frac{d}{d\tau} r_n - r_{n+1} - r_{n-1} + 2r_n -$$

$$- \frac{1}{2} [r_{n+1}q_{n+1} + r_{n-1}q_{n-1} + (2 - \delta_{n1} - \delta_{-n1}) r_n q_n] r_n = 0, \tag{12a}$$

$$r_n(\tau = 0) = (\delta_{ns} - \delta_{-ns}) \sqrt{g/2} \qquad (-\infty < n < \infty) \tag{12b}$$

on the infinite chain. This is connected with their equivalence, in the sense that the solutions coincide at $n \geq 1$. Let us pass over in (12a) to (12b) to a continuous descrip-

tion $r_n(\tau) \to r(n, \tau)$ and generalize, at the same time the form of the initial distribution assuming that it can extend over some molecules

$$i \frac{\partial}{\partial \tau} r(n, \tau) - \frac{\partial^2}{\partial n^2} r(n, \tau) - 2q(n, \tau)\, r^2(n, \tau) = 0 \ . \tag{13a}$$

$$r(n, 0) = \sqrt{\frac{g}{2l}} \begin{cases} 0, & -\infty \ < n < -s + \frac{1}{2} - l \ , \\ -1, & -s + \frac{1}{2} - l < n < -s + \frac{1}{2} \ , \\ 0, & -s + \frac{1}{2} \ < n < s - \frac{1}{2} \ , \\ 1, & s - \frac{1}{2} \ < n < s - \frac{1}{2} + l \ , \\ 0, & s - \frac{1}{2} + l \ < n < \infty \end{cases} \tag{13b}$$

$(s \geqq 1,\, l \geqq 1;\, -\infty < n < \infty) \ .$

So, we have arrived at the model of the nonlinear Schrödinger equation [8].

Information on the conditions of the formation and the most important physical parameters of solitons in such a model is involved in the transition coefficient $a^{-1}(\zeta)$ defined from the additional spectral problem [8] corresponding to the above model. Just the velocities $-4\xi_j$, amplitudes $2\eta_j$, and sizes $(2\eta_j)^{-1}$ of the exciting solitons

$$r_j^{(s)}(n, \tau \to \infty) = \frac{2\eta_j \exp\left[2i\xi_j n + 4i(\xi_j^2 - \eta_j^2)\,\tau + i\varphi_j\right]}{\text{ch}\left[2\eta_j(n + 4\xi_j\tau - m_j)\right]} \tag{14}$$

are given by the roots $\zeta_j = \xi_j + i\eta_j$ of the equation

$$a(\zeta) = 0 \qquad (\text{Im}\ \zeta > 0) \ . \tag{15}$$

If such roots do not exist, solitons are not generated and the initial distribution, in the long run, is dispersed over the whole chain.

It can be shown that in the case concerned (13b), (15) in notations

$$\zeta l = z \equiv -x + iy \ , \tag{16a}$$

$$g l/2 = G^2 \ , \tag{16b}$$

$$(2s - 1)/l = \Delta \tag{16c}$$

has the form

$$[(z^2 + G^2)^{1/2}\, \text{ctg}\, (z^2 + G^2)^{1/2} - iz]^2 + G^2 \exp\, (2iz\Delta) = 0 \quad (\text{Im}\ z > 0) \ . \tag{17}$$

The roots of this equation, if they do exist, appear in pairs $z_j^{\pm}(\Delta, G) = \mp\, x_j(\Delta, G) + iy_j(\Delta, G)$ $(x_j > 0,\, y_j > 0)$. However, to the real physical situation, i.e. solitons in a semi-infinite chain $(n \geqq 1)$, correspond only the roots with negative real part $z_j^{+}(\Delta, G)$, therefore these will be the subject of our interest.

Equation (17) generalizes naturally the equations obtained in other papers [5, 6] since, apart from the cases $\Delta = \infty$ and $\Delta = 0$, it allows one to consider uniquely the case of arbitrary Δ. Its analysis reveals that at each fixed Δ soliton formation is critical with respect to the parameter G. So a single soliton is formed in fulfilling the conditions

$$G_{\text{cr},1}(\Delta) < G \leqq G_{\text{cr},2}(\Delta) \ , \tag{18}$$

two solitons at

$$G_{\text{cr},2}(\Delta) < G \leqq G_{\text{cr},3}(\Delta) \ , \tag{19}$$

etc. The soliton velocities appear to be bounded,

$$x_j(\Delta, G) < x_{\text{cr},j}(\Delta) \ , \tag{20}$$

and each pair of values $G_{cr,j}(\varDelta)$ and $x_{cr,j}(\varDelta)$ $(j = 1, 2, 3, ...)$ is defined from the equation

$$(x_{cr}^2 + G_{cr}^2)^{1/2} \operatorname{ctg} (x_{cr}^2 + G_{cr}^2)^{1/2} = iG_{cr} \exp(-ix_{cr}\varDelta) - ix_{cr},$$

$$(G_{cr} > 0, \, x_{cr} > 0). \tag{21}$$

The functions $G_{cr,j}(\varDelta)$ and $x_{cr,j}(\varDelta)$ decrease monotonically with increasing $\varDelta$ so that

$$G_{cr,j}(0) = x_{cr,j}(0) = (j - \tfrac{1}{2}) \pi \sqrt{2}/2, \tag{22}$$

$$G_{cr,j}(\varDelta \gg 1) \approx (j - \tfrac{1}{2}) \pi/2, \tag{23a}$$

$$x_{cr,j}(\varDelta \gg 1) \approx (j - \tfrac{1}{2}) \pi/\varDelta. \tag{23b}$$

In particular, the dependences $G_{cr,1} = G_{cr,1}(\varDelta)$ and $x_{cr,1} = x_{cr,1}(\varDelta)$ are plotted in Fig. 1.

We note that at $\varDelta = 0$ the threshold value $G_{cr,1}(0)$ of the parameter G coincides with that quoted in Scott's paper [6]. At $\varDelta \gg 1$ the threshold value $G_{cr,1}(\varDelta \gg 1)$ of the parameter G proves to be smaller than the relevant threshold value $\pi/2$ for the infinite chain [1, 5, 6]. However, in fact, there is no contradiction since for any G lying in the interval $G_{cr,1}(\varDelta) < G \leqq \pi/2$ the value $y_1(\varDelta, G)$ with increasing $\varDelta$ tends to zero, and for $G > \pi/2$ to a certain finite value dependent on the difference $G - \pi/2$. The qualitative differences in the asymptotic $(\varDelta \gg 1)$ behaviour of $y_1(\varDelta, G)$ as a function of $\varDelta$ for $G \leqq \pi/2$ and $G > \pi/2$ can be clearly seen from Fig. 2.

For each fixed $\varDelta$ $y_1(\varDelta, G)$ is a monotonically increasing function of G, for example, for $\varDelta = 0$ we have

$$y_1(0, G) \approx G - G_{cr,1}(0), \qquad G - G_{cr,1}(0) \lessgtr G_{cr,1}(0). \tag{24}$$
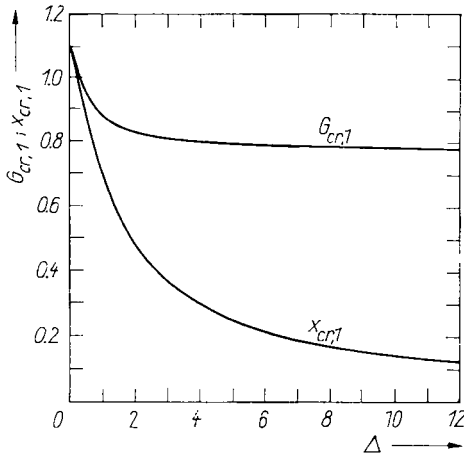

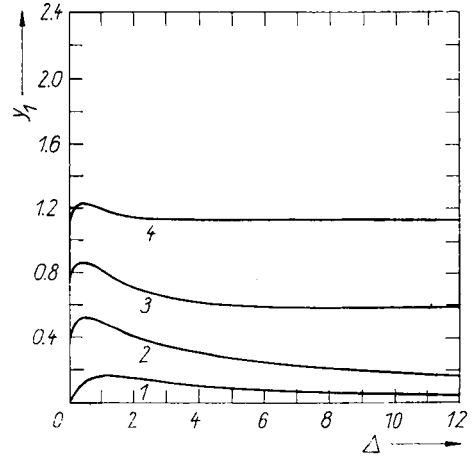Fig. 1   Fig. 2

Fig. 1. Dependences of the threshold nonlinearity parameter $G_{cr,1}$ and the limiting soliton velocity $v_{cr,1}$ on the inverse anisotropy parameter of the initial distribution $\varDelta$ $(v_{cr,1} = 4Ja^2x_{cr,1}/\hbar L$, a is the lattice constant, $L = la$ is the initial distribution size)

Fig. 2. Dependences of the inverse ratio of the characteristic size of the soliton to that of the initial distribution $2y_1$ on $\varDelta$ at different fixed values of the nonlinearity parameter G. (1) $G = \sqrt{2}\pi/4$, (2) $\pi/2$, (3) $5\pi/8$, (4) $3\pi/4$

However, the efficiency of soliton formation

$$D_1(\varLambda, G) \equiv \frac{\int\limits_0^\infty dn\, |r_1^{(s)}(n, \tau \to \infty)|^2}{\int\limits_0^\infty dn\, |r(n, 0)|^2} = 4y_1(\varLambda, G)/G^2 \tag{25}$$

depends on G in a non-monotonic way — at small values of $G - G_{cr,1}(\varDelta)$ it increases, and at large ones it decreases, so that at a certain optimal value $G_{max,1}(\varDelta)$ of the parameter G, the maximum part possible for a given $\varDelta$ of the initial distribution is transformed into a soliton (see Fig. 3).

It should be noticed that although the absolute efficiency maximum of soliton formation for the class of initial conditions under study is close to unity, it does not attain unity, i.e. part of excitation necessarily goes into soliton "tails". This is explained by the fact that even the minimum of the energy functional corresponding to a definite class of initial conditions normalized to one excitation cannot be lower than the energy of a single immobile soliton. As a result part of energy should be transferred to higher-energy delocalized "tail". If the given initial distribution provides no minimum of such a functional, the initial energy excess makes it possible to presume the formation of two and more solitons, since because of the binding energy loss the energy of the multisoliton state is always higher than that of single solitons. In the analysis of the initial distribution energy as a function of G the non-monotonic character of the G-dependent single-soliton formation efficiency also becomes clear. At small G the initial energy is positive and therefore solitons (bound states) cannot arise, the soliton formation efficiency equals zero. With increasing G the initial energy becomes negative and attains rapidly its minimum value which induces the conditions for soliton formation (18). Then the formation efficiency rapidly increases and reaches the maximum. Finally with further increase in the parameter G the initial energy increases slowly remaining negative. Therefore, the conditions (19) for the formation of several solitons are satisfied and the single-soliton formation efficiency decreases. The above-cited non-monotonic behaviour of the initial energy as a function of the parameter G is due to a competition between dispersion and binding energies in the initial distribution.

Besides the relationship between the initial distribution energy and the energies of one-, two-soliton, etc., states the result of the initial distribution evolution can essen-
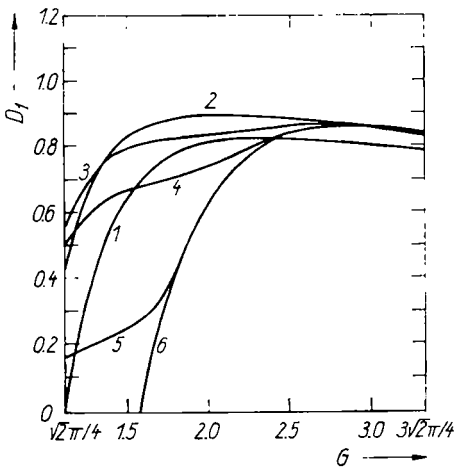

Fig. 3. Dependences of the soliton formation efficiency D_1 on the nonlinearity parameter G at different fixed values of $\varLambda$. (1) $\varDelta = 0$, (2) 0.4, (3) 1.0, (4) 2.0, (5) 12, (6) ∞
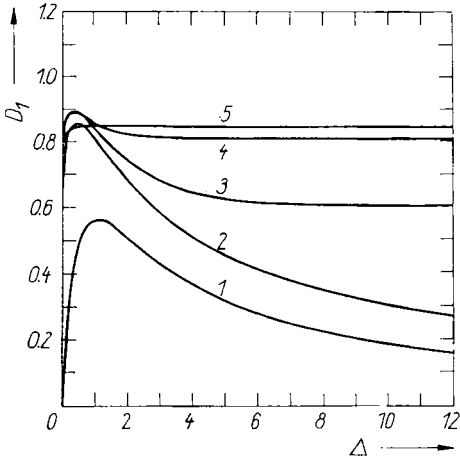
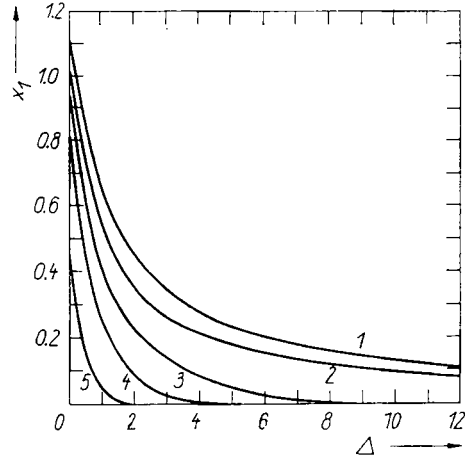Fig. 4   Fig. 5

Fig. 4. Dependences of the soliton formation efficiency D_1 on the inverse anisotropy parameter Δ at different fixed values of the nonlinearity parameter G. (1) $G = \sqrt{2}\pi/4$, (2) $\pi/2$, (3) $5\pi/8$, (4) $3\pi/4$, (5) $3\sqrt{2}\pi/4$

Fig. 5. Dependences of soliton velocity v_1 on the inverse anisotropy parameter Δ at different fixed values of the nonlinearity parameter G ($v_1 = 4Ja^2x_1/\hbar L$). (1) $G = \sqrt{2}\pi/4$, (2) $\pi/2$, (3) $5\pi/8$ (4) $3\pi/4$, (5) $3\sqrt{2}\pi/4$

tially be determined by the initial state anisotropy caused by the semi-infiniteness of the chain. This accounts for the fact that in the presence of the boundary the energy balance involves necessarily the kinetic energy of the evolution product motion deep into the chain. Such a motion arises due to wave reflection from the boundary and manifests itself with the growth of the parameter Δ^{-1}. Its physical meaning is evident (16c), with decreasing distance between the initial characteristic size of the initial distribution and the chain edge and/or with increasing characteristic size of the initial distribution the presence of the boundary should be pronounced.

Fig. 4 and 5 show how the soliton formation efficiency and also its velocity depend on the parameter Δ.
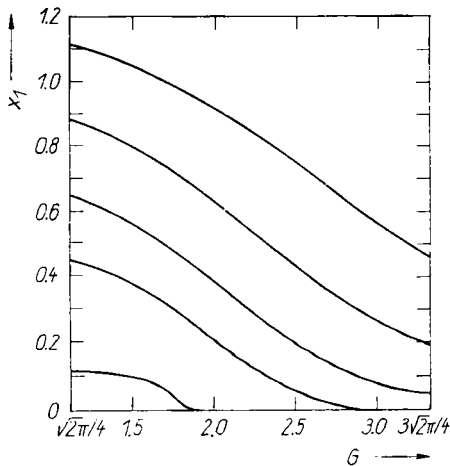

Fig. 6. Dependences of soliton velocity v_1 on the nonlinearity parameter G at different fixed values of the inverse anisotropy parameter Δ. (1) $\Delta = 0$, (2) 0.4, (3) 1.0, (4) 2.0, (5) 12.0 (numbered from the upper curve downward)

The relationship between soliton velocity and the parameter G follows from Fig. 6. With increasing G the soliton velocity decreases monotonically. If we here take into account the disregarded fact that a periodic potential appears on the way of soliton motion at large nonlinearity constant G [7], Hyman et al.'s result [9] on the possibility to localize the exciting soliton at the end of a discrete molecular chain becomes clear.

4. Concluding Remarks

In conclusion let us make some remarks. So to realize the soliton information transfer mechanism it seems to be of major importance to govern the soliton velocity and the efficiency of its generation by changing the input place and the localization degree of the exciting signal. One should have, however, in mind that when the exciting signal input is at large distances from the chain edge the soliton mechanism for transport can be difficult because of the pinning of slow solitons conditioned by the discreteness of the chain.

Attention should also be paid to the fact of the higher than pointed out ealier [5, 6] soliton formation efficiency at smaller nonlinearity constant (compare curve 2 with curves 1 and 6 in Fig. 3).

At last we should indicate the possibility of some change in the criteria (obtained in this paper) of soliton formation with account of energy dissipation. In particular, it seems quote plausible that under certain conditions these criteria will turn out to be less rigid since the dissipative channel can carry away the initial distribution of energy excess, which in absence of dissipation can be spent to generate soliton "tails" or soliton satellites. This interesting problem requires still its final solution.

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